IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants:	Haruyuki MAKIO et al.	)	Confirmation No.8543
Application No.:	10/569,475	)	Art Unit: 1796
Filed:	February 24, 2006	)	Examiner: O. Asinovsky
Title:	TELECHELIC POLYOLEFIN	)	

DECLARATION UNDER 37 C.F.R. §1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Dear Sir/Madam:

I, Nobuo Kawahara hereby declare as follows:

1. I am a graduate of Shizuoka University, receiving a master's degree in engineering from Shizuoka University in 1993 and doctor's degree in engineering from Shizuoka University in 2007.

2. In 1993, I joined Mitsui Toatsu Chemicals (Current, Mitsui Chemicals, Inc.) and have been working as a researcher since 1993 (from 2002 as a senior researcher). My research fields have been covered olefin polymerization catalysts and a creation of functional polymer materials. My research theme and research results including publications and other activities are listed in an attached file as attachment 1.

3. I have read and am familiar with the subject matter of the above-identified patent application. I have carried out the following experiments in order to demonstrate to clear the difference of the polymers disclosed in U. S. Patent 6,274,688 (hereinafter Nakagawa) and those of the present invention.

4. Olefin polymerization was carried out with an atom transfer radical polymerization (hereinafter ATRP) method according to Nakagawa. More specifically, polymerization was carried out under conditions with and without n-butylacrylate as comonomer.

Experiments 1 to 8 were carried out by a scheme shown in Appendix 1. Samples obtained were analyzed by ^{1}H-NMR measurement and GPC measurement. The ^{1}H-NMR chart and GPC chart obtained by the measurement are shown in Appendix 2 and appendix 3, respectively. The results of these experiments are shown below.

Results of ATRP of n-BuA

Exp.#	nBuA[a)]		EA[b)]	Olefin feed		Yield[c)]	GPC[d)]			Olefin Contents[e)]
	(ml)	(mmol)	(ml)			(g)	Mw (g/mol)	Mn (g/mol)	Mw/Mn	
1	100	698	0	none		87.3	18300	13900	1.3	none
2	10	70	90	none		10.3	2490	1670	1.5	none
3	10	70	90	Ethylene	(30NL/h)	10.1	2310	1460	1.6	none (or trace?)
4	0		100	Ethylene	(30NL/h)	0	-	-	-	-
5	10	70	90	Propylene	(30NL/h)	9.7	2220	1390	1.6	none (or trace?)
6	0		100	Propylene	(30NL/h)	0	-	-	-	-
7	50	349	0	Octene-1	(50ml)	58.1	11500	6160	1.9	ca. 35 mol%
8	0		0	Octene-1	(100ml)	0	-	-	-	-

Polymerization condition; CuBr (5 mmol), PMDETA (5 mmol), Initiator (5 mmol), 70 °C, 4 hr.
a) n-Butyl acrylate, b) Ethyl acetate, c) included residual materials (catalyst, solvent, monomer, etc),
d) PS standard, e) estimated by ^{1}H NMR

Details of each experiment are described as follows.

[Experiment 1] Homo-polymerization of n-butylacrylate by an ATRP method

Into a glass reactor having a volume of 500 mL equipped with a stirrer, a condenser tube, a glass inlet tube and a thermocouple, n-butylacrylate (100mL) was added and the system was purged sufficiently with nitrogen for two hours, then the content was heated to 70°C with stirring. Thereto were added a solution of ethyl-2-bromobutylate/acetonitrile (1M; 5mL) and a mixture of copper bromide (I) (5mmol), pentamethyldiethylenetriamine (5mL) and acetonitrile (5mL), then heated with stirring for 4 hours at 70°C. Then introducing oxygen to the system, the reactor was cooled in an ice bath. Solvent and residual monomer was removed by evaporation to obtain 87.3 gram of viscous liquid. From GPC analysis, weight average molecular weight (Mw) and number average molecular weight (Mn) of the obtained liquid was determined to be 18,300 and 13,900

respectively. NMR chart and GPC chart of the liquid are shown in Figure 2 of Appendix 2 and Figure 1 of Appendix 3, respectively.

[Experiment 2] Homo-polymerization of n-butylacrylate by an ATRP mcthod under a presence of solvent

The same procedure was carried out as the experiment 1, except that ethyl acetate (90mL) and n-butylacrylate (10mL) was added to the glass reactor to obtain 10.3 gram of viscous liquid. From GPC analysis, weight average molecular weight (Mw) and number average molecular weight (Mn) of the obtained liquid was determined to be 2,490 and 1,670 respectively. NMR Chart and GPC chart of the liquid are shown in Figure 3 of Appendix 2 and Figure 2 of Appendix 3.

[Experiment 3] Co-polymerization of n-butylacrylate with ethylene by an ATRP method under a presence of solvent

Into a glass reactor having a volume of 500 mL equipped with a stirrer, a condenser tube, a glass inlet tube and a thermocouple, ethyl acetate (90mL) and n-butylacrylate (10mL) were added and the system was purged sufficiently with nitrogen for two hours, then the content was heated to 70°C with stirring while introducing ethylene to the system (30NL/h). Thereto were added a solution of ethyl-2-bromobutylate/acetonitrile (1M; 5mL) and a mixture of copper bromide (I) (5mmol), pentamethyldiethylenetriamine (5mL) and acetonitrile (5mL), then heated with stirring for 4 hours at 70°C while introducing ethylene (30NL/h). Then introducing oxygen to the system, the reactor was cooled in an ice bath. Solvent and residual monomer was removed by evaporation to obtain 10.1 gram of viscous liquid. From GPC analysis, weight average molecular weight (Mw) and number average molecular weight (Mn) of the obtained liquid was determined to be 2,310 and 1,460 respectively. MNR chart and GPC chart of the liquid are shown in Figure 4 of Appendix 2 and Figure 3 of Appendix 3, respectively. Olefin was not detected in the liquid by NMR analysis.

[Experiment 4] Homo-polymerization of ethylene by an ATRP method under a presence of solvent

Into a glass reactor having a volume of 500 mL equipped with a stirrer, a condenser tube, a glass inlet tube and a thermocouple, ethyl acetate (90mL) was added and the system was purged sufficiently with nitrogen for two hours, then the content was heated to 70°C with stirring while introducing ethylene to the system (30NL/h). Thereto were added

a solution of ethyl-2-bromobutylate/acetonitrile (1M; 5mL) and a mixture of copper bromide (I) (5mmol), pentamethyldiethylenetriamine (5mL) and acetonitrile (5mL), then heated with stirring for 4 hours at 70°C while introducing ethylene (30NL/h). Then introducing oxygen to the system, the reactor was cooled in an ice bath. Solvent and residual monomer was removed by evaporation to obtain catalyst residue without polymer.

[Experiment 5] Co-polymerization of n-butylacrylate with propylene by an ATRP method under a presence of solvent

The same procedure was carried out according to the experiment 3, except that propylene (30NL/h) was introduced to the glass reactor instead of ethylene to obtain 9.7 gram of viscous liquid. From GPC analysis, weight average molecular weight (Mw) and number average molecular weight (Mn) of the obtained liquid was 2,220 and 1,390 respectively. NMR chart of the liquid is shown in Figure 5 of Appendix 2. Olefin was not detected in the liquid by NMR analysis.

[Experiment 6] Homo-polymerization of propylene by an ATRP method under a presence of solvent

The same procedure was carried out according to the experiment 4, except that propylene (30nL/h) was introduced to the glass reactor instead of ethylene to obtain no polymer.

[Experiment 7] Co-polymerization of n-butylacrylate with octene by an ATRP method

Into a glass reactor having a volume of 500 mL equipped with a stirrer, a condenser tube, glass inlet tube and a thermocouple, n-butylacrylate (50mL) and 1-octene (50mL) were added and the system was purged sufficiently with nitrogen for two hours, then the content was heated to 70°C with stirring. Thereto were added a solution of ethyl-2-bromobutylate/acetonitrile (1M; 5mL) and a mixture of copper bromide (I) (5mmol), pentamethyldiethylenetriamine (5mL) and acetonitrile (5mL), then heated with stirring for 4 hours at 70°C. Then introducing oxygen to the system, the reactor was cooled in an ice bath. Solvent and residual monomer was removed by evaporation to obtain 48.1 gram of viscous liquid. From GPC analysis, weight average molecular weight (Mw) and number average molecular weight (Mn) of the obtained liquid was 11,500 and 6,160 respectively. NMR chart and GPC chart of the liquid are shown in Figure 6 of Appendix 2 and Figure 5 of Appendix 3, respectively. The amount of octene in the polymer was calculated to be 35% by mole by NMR analysis.

[Experiment 8] Homo-polymerization of 1-octene by an ATRP method

Into a glass reactor having a volume of 500 mL equipped with a stirrer, a condenser tube, a glass inlet tube and a thermocouple, 1-octene (100mL) was added and the system was purged sufficiently with nitrogen for two hours, then the content was heated to 70°C with stirring. Thereto were added a solution of ethyl-2-bromobutylate/acetonitrile (1M; 5mL) and a mixture of copper bromide (I) (5mmol), pentamethyldiethylenetriamine (5mL) and acetonitrile (5mL), then heated with stirring for 4 hours at 70°C. Then introducing oxygen to the system, the reactor was cooled in an ice bath. Solvent and residual monomer was removed by evaporation to obtain catalyst residue without polymer.

Results

(1) No polymer was obtained in cases of homo-polymerization by an ATRP method (Experiment 4, Experiment 6 and Experiment 8).

(2) Butylacrylate homo-polymer was obtained in case of co-polymerization with ethylene or propylene (Experiment 3 and Experiment 5).

(3) n-butylacrylate/1-octene copolymer was obtained in a case of co-polymerization of 1-octene with n-butylacrylate by a method of ATRP. However 1-octene content of the copolymer was as low as about 35% by mole and molecular weight distribution, represented by Mw/Mn was increased to around a little bit lower than 2 (Experiment 7).

Conclusion

From the results described above, it is impossible to homo-polymerize non-polar olefin monomer by an ATRP method, which is disclosed in Nakagawa. Regarding a copolymerization of non-polar olefin monomer with acryl monomer, the result shows that copolymer having non-polar olefin unit in the copolymer cannot be obtained or copolymer having non-polar olefin unit in the copolymer may be a minor component (less than 50% by mole) having broader molecular weight distribution, even if it is obtained.

Consequently, the judgment of the examiner that the polyolefin of the present invention is disclosed in Nakagawa is not correct; it is also clear from a description of the document (Principles of Polymerization (1991), which the applicant submitted in the response to the Office Action dated April 3, 2008.

Therefore the present invention could not be rejected being anticipated by Nakagawa or being obvious over Nakagawa in view of other prior arts.

5. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment or both, under

Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

Nobuo Kawahara

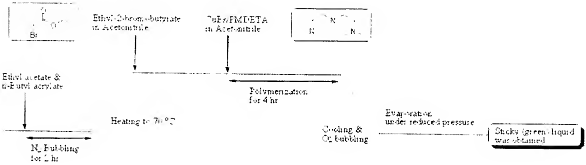
Nobuo Kawahara

April 28, 2009

Date

Appendix 1

Scheme in a case of Experiment 2

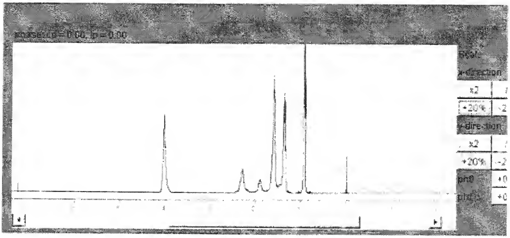


Appendix 2

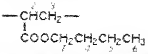
Figure 1: Estimated NMR chart of Poly(N-Butylacrylate)

^{1}H 500 MHz NMR Spectra for poly(butyl acrylate) [PID: P040006]

- Sample name: Poly(butyl acrylate)
- Measurement conditions:
  - Solvent: $CDCl_3$
  - Concentration: 5 %[wt/vol]
  - Temperature: 60 °C
  - ^{1}H NMR Standard: TMS
  - Pulse interval: 3 sec
  - Scan number: 256



- Peak assignment:



chemical shift (ppm)	Assignment
4.04	1
2.30	2
1.91	[illegible] (m)
1.61	4
1.48	3a (m)
1.38	5
0.93	[illegible]

[Note for Assignment]

$CHCl_3$:7.28ppm

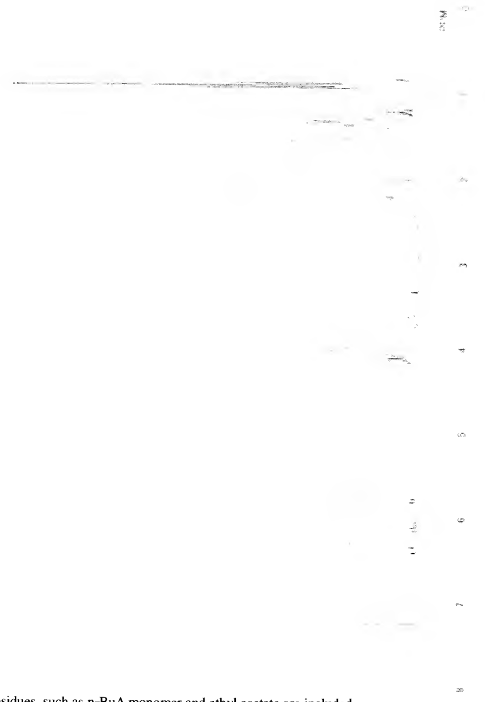
Appendix 2 (continued)

Figure2: NMR chart of a sample obtained by experiment 1

* Residues, such as n-BuA monomer and ethyl acetate are included

Appendix 2 (continued)

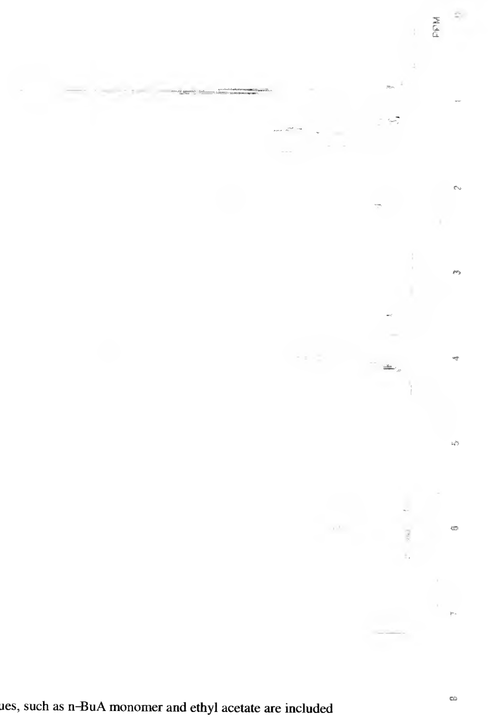
Figure3: NMR chart of a sample obtained by experiment 2



* Residues, such as n-BuA monomer and ethyl acetate are included

Appendix 2 (continued)

Figure 4: NMR chart of a sample obtained by experiment 3



Residues, such as n-BuA monomer and ethyl acetate are included

Appendix 2 (continued)

Figure 5: NMR chart of a sample obtained by experiment 5

Residues, such as n-BuA monomer and ethyl acetate are included

Appendix 2 (continued)

Figure 6: NMR chart of a sample obtained by experiment 7

Residues, such as n-BuA monomer and ethyl acetate are included

Appendix 3

Figure 1: GPC chart of a sample obtained by Experiment 1

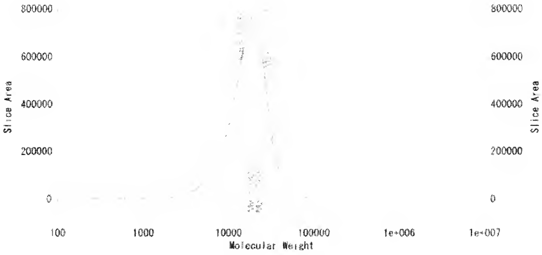
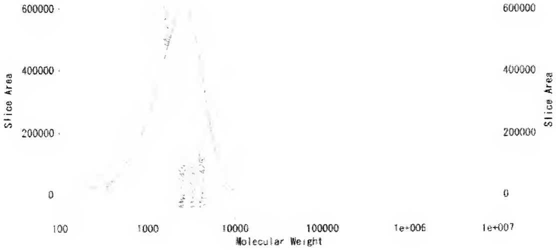


Figure 2: GPC chart of a sample obtained by Experiment 2



Appendix 3 (continued)

Figure 3: GPC chart of a sample obtained by Experiment 3

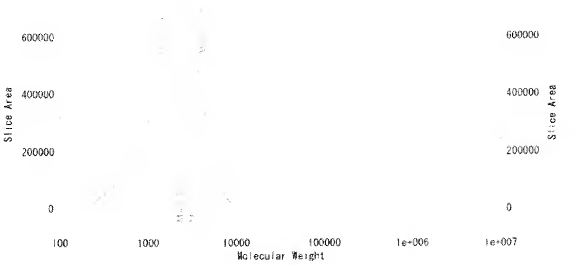
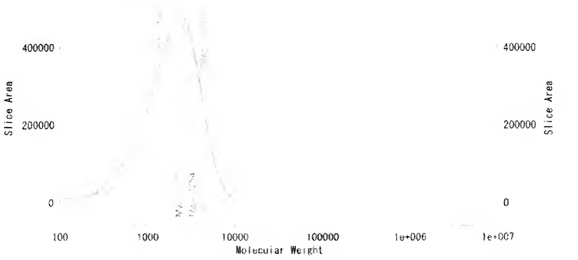
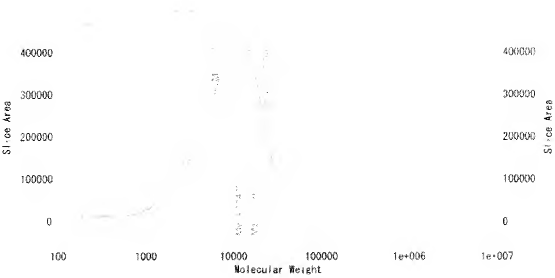


Figure 4: GPC chart of a sample obtained by Experiment 5



Appendix 3 (continued)

Figure 5: GPC chart of a sample obtained by Experiment 7



Attachment 1

Research theme and research results of Dr. N. Kawahara

Papers (selected)

1) H. Kaneko, J. Saito, N. Kawahara, S. Matsuo, T. Matsugi, N. Kashiwa:
"Synthesis and characterization of polypropylene-based block copolymers possessing polar segments via controlled radical polymerization"
Journal of Polymer Science: Part A: Polymer Chemistry, Vol.47, pp812-823(2009).

2) N. Kawahara, J. Saito, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
"Polymer hybrids based on polyolefins – syntheses, structures and properties"
Advances in Polymer Science, Vol.217, pp79-119(2008).

3) H. Kaneko, J. Saito, N. Kawahara, S. Matsuo, T. Matsugi, N. Kashiwa:
"Synthesis and characterization of polypropylene-based polymer hybrids linking poly(methyl methacrylate) and poly(2-hydroxyethyl methacrylate)"
Polymer, Vol.49, pp4576-4584(2008).

4) N. Kawahara, J. Saito, S. Matsuo, H. Kaneko, T. Matsugi, Y. Toda, N. Kashiwa:
"Study on unsaturated structures of polyhexene, poly(4-methylpentene) and poly(3-methylpentene) prepared with metallocene catalysts"
Polymer, Vol.48, pp425-428(2007).

5) N. Kawahara, J. Saito, S. Matsuo, H. Kaneko, T. Matsugi, S. Kojoh, N. Kashiwa:
"New methodology for synthesizing polypropylene-graft-polystyrene (PP-g-PS) by coupling reaction with brominated polypropylene"
Polymer Bulletin, Vol.59, pp177-183(2007).

6) H. Kaneko, S. Matsuo, N. Kawahara, J. Saito, T. Matsugi, N. Kashiwa:
"Synthesis and mechanical properties of polypropylene-based polymer hybrids via controlled radical polymerization"
Macromolelular Symposia, Vol.260, pp9-14(2007).

7) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T. Matsugi, J. Saito, N. Kashiwa:
"Synthetic method of polyethylene-poly(methylmethacrylate) (PE-PMMA) polymer hybrid via reversible addition-fragmentation chain transfer (RAFT) polymerization with functionalized polyethylene"
Polymer Bulletin, Vol.57, pp805-812(2006).

8) H. Kaneko, T. Matsugi, N. Kawahara, S. Matsuo, S. Kojoh, N. Kashiwa:
"Polypropylene-*block*-poly(methylmethacrylate) and -*block*-poly(*N*-isopropylacrylamide) block copolymers prepared by controlled radical polymerization with polypropylene macroinitiator"
Kinetics and Catalysis, Vol.47, pp227-233(2006).

9) S. Matsuo, T. Matsugi, J. Saito, N. Kawahara, H. Kaneko, N. Kashiwa:
"Creation of new polyolefin hybrids on the surface of molded polypropylene sheet"
Studies in surface science and catalysis, Vol.161, pp1-6(2006).

10) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
"Investigation of insertion reaction of 10-undecen-1-ol protected with alkylaluminum in $En(Ind)_2ZrCl_2$/MAO catalyst system"
Journal of Molecular Catalysis A: Chemcal, Vol.241, pp156-161(2005).

11) H. Kaneko, S. Kojoh, N. Kawahara, S. Matsuo, T. Matsugi, N. Kashiwa:

"Syntheses of graft and star copolymers possessing polyolefin branches by using polyolefin macromonomer"
Journal of Polymer Science: Part A: Polymer Chemistry, Vol.43, pp5103-5118(2005).

12) N. Kawahara, S. Kojoh, Y. Toda, A. Mizuno, N. Kashiwa:
"The detailed analysis of the vinylidene structure of metallocene-catalyzed polypropylene"
Polymer, Vol.45, pp355-357(2004).

13) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T. Matsugi, Y. Toda, A. Mizuno, N. Kashiwa:
"Study on chain end structures of polypropylenes prepared with different symmetrical metallocene catalysts"
Polymer, Vol.45, pp2883-2888(2004).

14) H. Kaneko, S. Kojoh, N. Kawahara, S. Matsuo, T. Matsugi, N. Kashiwa:
"Polymacromonomers with polyolefin branches synthesized by free radical homopolymerization of polyolefin macromonomer with a methacryloyl end group"
Macromolelular Symposia, Vol.213, pp335-345(2004).

15) N. Kashiwa, S. Kojoh, N. Kawahara, S. Matsuo, H. Kaneko, T. Matsugi:
"New methodology for synthesizing polyolefinic graft block copolymers and their morphological features"
Macromolecular Symposia, Vol.201, pp319-326(2003).

16) N. Kashiwa, T. Matsugi, S. Kojoh, H. Kaneko, N. Kawahara, S. Matsuo, T. Nobori, J. Imuta:
"Functionalization of polyethylene based on metallocene catalysis and its application to syntheses of new graft copolymers possessing polar polymer segments"
Journal of Polymer Science: Part A: Polymer Chemistry, Vol.41, pp3657-3666(2003).

17) T. Matsugi, S. Kojoh, N. Kawahara, S. Matsuo, H. Kaneko, N. Kashiwa:
"Synthesis and morphology of polyethylene-block-poly(methylmethacrylate) through the combination of metallocene catalysis with living radical polymerization"
Journal of Polymer Science: Part A: Polymer Chemistry, Vol.41, pp3965-3973(2003).

Oral and Poster Presentations (selected)

1) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T Matsugi, J. Imuta, N. Kashiwa:
MetCon2003, Session III, Houston (2003).
2) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
40th IUPAC World Polymer Congress, P2.1-74, Paris (2004).
3) N. Kawahara, S. Kojoh, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
International Chemical Congress of Pacific Basin Societies, 1024, Hawaii (2005).
4) J. Saito, N. Kawahara, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
1st European Chemistry Congress, M-OC-108, Budapest (2006).
5) N. Kawahara, J. Saito, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
234th ACS National Meeting, PMSE0043, Boston (2007).
6) N. Kawahara, J. Saito, S. Matsuo, H. Kaneko, T. Matsugi, N. Kashiwa:
FlexPO 2008, Session II, Houston/Galveston (2008).

DECLARATION

I, Makoto TAKEZAWA, c/o S.SUZUKI & ASSOCIATES, 13-6 Nishigotanda 7-chome, Shinagawa-ku, Tokyo, JAPAN, do solemnly and sincerely declare that I understand the Japanese language and the English language well, and that the attached English version is a full, true and faithful translation made by me of the Japanese Patent Application No. 2003-302240 filed on August 27, 2003.

I make this solemn declaration conscientiously believing the same to be true.

March 26, 2009

Makoto Takezawa

Makoto TAKEZAWA